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SOURCE Izvestiya Akademii Nauk SSSR, Otdeleniye Khimicheskikh Nauk, No 2, 1950.

SOVIET SCIENTISTS ON ACHIEVEMENT OF EQUILIBRIUM IN DISTRIBUTION OF A MICROCOMPONENT BETWEEN SOLID CRYSTALLINE ISOMORPHOUS PHASE AND MELTED SALT

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[A Digest]

In prior work by Khlopin and Klokman, Izvestiya Akademii Nauk SSSR, Otdeleniye Khimicheskikh Nauk, No 3, 254, 1949, it was shown that equilibrium distribution of radium between crystals of salts isomorphous with it and a liquid melt containing the same salts is reached very rapidly (within 2-4 hours) starting from either side, irrespective of energetic agitation or the absence of it. There is a sharp difference between this process and distribution between crystals of the macrocomponent and a solution not only with respect to speed, but also as far as the type of distribution is concerned. While Doerner's and Hoskins' logarithmic law applies in the case of a solution, Berthelot-Nernst-Khlopin's law was found to apply in the case under consideration. According to Doerner's and Hoskins' law, distribution of radium could not be observed whenever the solid phase of the macrocomponent was obtained by crystallization from melts which remained in equilibrium with the crystals of the macrocomponent.

The suspicion arose that diffusion plays the main role in establishing an adsorption equilibrium under the conditions existing here, and this matter was checked experimentally on the system

$$Ba(NO_3)_2 - Ra(NO_3)_2 - NaNO_3$$

which had already been studied in an earlier work. It could be established that in counterdistinction to conditions obtaining in the solution system, the speed with which equilibrium distribution of the microcomponent was attained in crystals melted at 315 degrees centigrade appeared to be independent of the rate of agitation.

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Using the relationships derived by A. N. Murin (A. E. Polesitskiy and A. N. Murin, Doklady Akademii Nauk SSSR, Vol XIV, 6, 1944), the apparent rate of diffusion could be calculated from the observed speed with which equilibrium was established in this case. By using crystals of $Ba(NO_3)_2$ which were 100 times as large as those used in the original series of experiments (average diameter of 2.6×10^{-2} as compared with 2.9×10^{-4} centimeters), the time necessary for attaining the equilibrium was multiplied by a factor of two, reaching the value of 8 hours. If equilibrium were established as a result of diffusion in the solid phase, the time would have increased by a factor of 10^4 rather than 2.

An error could have occurred in estimating the dimensions according to the apparent diameter determined microscopically. This could have been due either to the presence of surface cracks (in the case of the 2.9×10^{-4} centimeter crystals) or the adhesion of smaller crystals to the surface (in the case of the 2.6×10^{-2} centimeter crystals). The first possibility was excluded by adsorbing ThX and estimating the diameter of the crystals on the basis of the surface determined radiometrically by comparison with well-defined $Ba(NO_3)_2$ crystals of the size 200-210 μ which had also been treated the ThX. The diameter obtained in this manner was found to be 2.9 μ and, consequently, checked with that determined microscopically. The second source of error was excluded as far as possible by letting the $Ba(NO_3)_2$ crystallize very slowly, sifting, heating to 315 degrees, sifting again, and finally washing very thoroughly with alcohol. The diameter of the crystals obtained in this manner was found to be 5.4×10^{-2} centimeters by microscopic measurement. The time required for attaining a state of adsorption equilibrium was only 15 hours in the case of these crystals, i.e., it was much smaller than that which would have been required if equilibrium were achieved by diffusion in the solid phase. The apparent coefficient of diffusion, obtained on the basis of experimental data, was 1.2×10^{-4} centimeters square per day in this instance as compared with 1.03×10^{-8} centimeters square per day in the 2.9×10^{-4} centimeter crystals.

These results show that equilibrium with respect to adsorption of the microcomponent is not reached as a result of diffusion in the solid phase, but by virtue of recrystallization. Why, then, is equilibrium attained so rapidly and at a rate which is independent of the speed of agitation? It appears that equalization of concentration in melts is expedited by convection currents and that diffusion of ions in melts is very rapid due to the absence of a hydrate sheath.

The fact that there is no appreciable diffusion of radium or its isotope ThX in crystals of barium nitrate could be verified by treating barium nitrate crystals with a radium or ThX solution in 96 percent alcohol and determining the amount of radon or thoron emitted by the dried crystals after the latter had been kept at a certain temperature (temperature of diffusion) for a certain length of time (time of diffusion). This is essentially the method used by O. Hahn, but his technique was improved by carrying out the measurements on crystals with a surface approaching the ideal, so that the emanating capacity was no less than 40-41 percent at room temperature. By this method. the coefficient of diffusion was found to be $D_{Ra}^{300} = 1 \times 10^{-12}$ centimeters square per day, showing that diffusion is negligible and cannot be held responsible for establishing the equilibrium of radium between the solid phase and the melt.

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